AD NUMBER
AD362715

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31 Jul 1977, DoDD 5200.10; NOTS ltr Jul 1978

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ELECTRON ACCEPTORS AS ELECTRON SCAVENGERS
IN THE IONOSPHERE

(U)

by

P. R. Hammond

Research Department

U.S. NAVAL ORDNANCE TEST STATION

China Lake, California

July 1965

U. S. NAVAL ORDNANCE TEST STATION

AN ACTIVITY OF THE BUREAU OF NAVAL WEAPONS

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FOREWORD

This work was started September 1964 at the University of Leicester, England, where the author held the post of ICI Fellow. It was continued at the U. S. Naval Ordnance Test Station under Foundational Research, Bureau of Naval Weapons Task Assignment R360-FR 106/216-1/R011-01-01 from December 1964 to April 1965. It is transmitted for information only and does not necessarily represent the official views or final judgment of the Station.

ABSTRACT

It is proposed that molecules with high
electron affinity, such as carbonyl cyanide
and tetracyanoethylene, should be discharged
into the ionosphere, so scavenging areas of
this region of electrons. Electron capture
will produce radical anions and other chemical
products of the acceptors, hence the electri-
cal properties of the area will be altered
drastically. There will be a temporary effect
on radio-wave propagation and reflection.
Classes of electron accepting chemicals are
proposed. The results of the scavenging
experiments of Project Firefly are briefly
discussed. (CONFIDENTIAL)

ACKNOWLEDGMENT

CONTENTS

ELECTRON DENSITY IN THE IONOSPHERE

Ionization of the upper levels of the atmosphere is produced by X-ray, ultraviolet, and cosmic radiation. To some extent there is a selective absorption of this radiation at different levels, depending on the density and chemical composition of the air, and equilibria are set up where the rate of dissociation to electrons and cations is balanced by the rate of recombination of positively and negatively charged species. Density distribution, n_e in electrons cm^{-3} as examined by radio-wave propagation (Ref. 1 and 2) and rocket-borne experiments (Ref. 3), roughly follows a layer pattern. These are the D-layer, 60-85 km, electron density maximum around 10^3 electrons cm^{-3}; the E-layer, 85-140 km, 10^5 electrons cm^{-3}; and the F-layer, which for the purposes of this proposal may be considered from 140-500 km, 10^6 electrons cm^{-3}. The maximum densities quoted are for normal daytime conditions. They depend on latitude, and decrease at night by as much as an order of magnitude. In addition, changes of height of the layer maxima take place, whereas splitting into further layers, particularly the F, occurs. Solar storms alter the distribution greatly, causing an increase by as much as two orders of magnitude in the D and E layers. High altitude atomic explosions also change the density distribution.

A distribution ratio of the total number of electrons for what may well be typical daytime conditions is $1:10^2:5 \times 10^3$ for the D:E:F layers, with a combined density of 2×10^{13} electrons cm^{-2} of the earth's surface up to a height of 500 km.

For the purpose of this proposal, removal of electrons from a volume contained within a 100-km square on the ground to a height of 500 km is considered. Such a volume, which contains approximately 2×10^{27} electrons or 3×10^3-gram equivalents of electrons, should be sufficient for a definitive experiment on radio-wave propagation.

For example, just matching every electron in the volume with 1 atom of fluorine will require 60 kg of this element. Extending the calculation to the whole of the earth's ionosphere for the 2×10^{13} electrons cm^{-2} figure requires 3,000 long tons of fluorine, and not 50 as quoted in the literature (Ref. 4).

EFFECTS OF ELECTRON REMOVAL

Electrons in the ionosphere have three effects on a radio wave: increase of velocity and hence a decrease of refractive index; absorption, arising from collision of the oscillating electrons with gas molecules; and polarization. Diminishing the number of electrons will diminish these effects.

The refractive index decreases with increasing n_e and with increasing wavelength. The decreasing refractive index with height to a minimum in the F-layer, produces reflection of intermediate frequency rays back to the earth's surface. For any particular frequency, a layer exists, where the more oblique rays are reflected and the steeper pass through. Although reception is strong near the transmitter or at the point where the reflected rays return to earth, it is very weak in between. For high frequencies, there are no regions of sufficiently large n_e to cause reflection. For low frequencies, even the low density D-layer is effective.

For intermediate frequencies, where there is reflection from the E- and F-layers, attenuation occurs in the D-layer, increasing with increasing n_e and with increasing molecular density. Replacing the free oscillating electrons here with negative ions, caused by the addition of a scavenger, should improve long-distance radio communication. Alternatively, very low frequency transmission as used for the fleet communications network should be hampered if electrons were captured in this layer above the radio station. Although the chemically reactive acceptor molecules will be steadily destroyed in the atmosphere of the D-layer, many are available of much greater stability than the alkali metals (Ref. 4) at this height. These include most of the acceptors that are listed in Table 1 in the next section.

At a critical wavelength, dependent on the electron density distribution, there is reflection from the high and most concentrated n_e zones of the F-layer and only at very oblique angles of incidence. This occurs in the 10-meter wavelength region, and is the basis of a long-range radar detection probe (Ref. 5). Interference with the probe is the main concern of this proposal. In operation, the waves are sent at a glancing angle to the horizon towards an enemy installation, and are reflected from the ionosphere to earth at a distance, approximately 2,000 miles, depending on the height used for reflection. A moving object reflects back waves, at a slightly displaced frequency, to the ionosphere and then to a receiver at the position of the transmitter. The system is blind to the area of the earth's surface over the horizon to the point where the reflected ray returns. Replacement of negative ions for electrons at the point in the ionosphere where reflection takes place should effectively disarm the probe.

ELECTRON ACCEPTORS[1]

Electron Affinity. In the low pressure of the F-layer of the ionosphere, three electron capturing processes will be possible--radiative attachment, nonradiative attachment, and dissociative attachment. Success in electron scavenging and, hence, success of the project relies chiefly on high electron affinities, E, of the acceptor molecules. The act of capturing an electron may be exothermic up to the adiabatic value of E. If E is large, the excited radical anion is likely to be short-lived and may loose this energy by radiation. Alternatively a multi-atomic acceptor may accommodate vibrational energy or may rupture.

Very few measurements indeed have been reported of the electron affinities of molecules. (1) Isolated compounds are adapted to Born-Haber cycle calculations (Ref. 6 and 7). (2) The few techniques aimed at measuring E directly are pioneering in nature and as such must be viewed skeptically at present (Ref. 8-12). (3) Quite reliable information on the relative values of electron affinity certainly for organic compounds come from charge-transfer (Ref. 13) and polarographic (Ref. 14) studies. (4) Strong electron acceptors are reactive chemically, exhibiting electrophilic, Lewis acid, and oxidizing character.
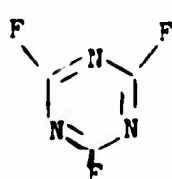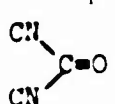
Selection of a compound as an electron acceptor must be based therefore on a detailed examination of its physical and chemical properties. For this reason, the strength of acceptors listed in Table 1 are classified in a qualitative manner, mainly on the considerations above but in part on an unavoidable intuitive analysis, as strong (S), medium (M), and weak (W). Very approximately, strong corresponds to electron affinities in the region 2-4 ev, medium 1-2 ev, and weak 0-1 ev.

Classes of Compounds. Table 1 stresses mainly organic acceptors, although a few inorganic compounds are also included. Radical cation and radical acceptors, e.g., N_2, are omitted. An interesting possibility would be if the latter could be discharged into the ionosphere as a result of incomplete combustion--either of the payload, or of an appropriately designed rocket fuel.

Table 1 is by no means an exhaustive list of possible scavengers, but rather a representative of the class of materials. The compounds selected are reported in the chemical literature, many more can be visualized. The choice of compounds shown arises from a compromise among molecular weight, acceptor strength, volatility, and chemical stability. It is of interest that many of the recommended scavengers have been studied by the Central Research Laboratories of the E. I. Du Pont Chemical Company, Wilmington, Delaware.
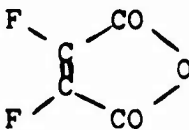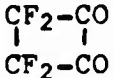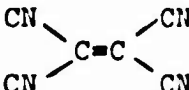
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[1] This portion of the report is UNCLASSIFIED.

TABLE 1. Proposals for Electron Accepting Chemicals

Formula	Mol. wt.	Acceptor strength[a]	Volatility,[b] °C	Chemical stability[c]	Recom- mended
Oxygen	32	W	b.p. -183	S	..
Halogens	38-254	M	from b.p. -188 to readily s.	S	R
NO_2	46	S	N_2O_4 m.p. -9.3, b.p. 21.3	S store as N_2O_4, decomposes to NO_2 in vapor	R
SO_2	64	M	b.p. -10	S	R
High valency fluorides e.g., SbF_5	217	S	m.p. 7, b.p. 150	S	..
BF_3	68	M	b.p. -101	S	R
$Cl-C\equiv N$	61.5	W	m.p. -6, b.p. 14	S on careful storage	..
$F-C\equiv N$	45	W	s. -72	S on careful storage	..
CF_2O	66	W	b.p. -83	S	..
$CF_2=NF$	83	M	b.p. -101	U	..
CF_3NO_2	115	W	b.p. -31	P	..
CNCOF	73	M	b.p. -20	P	P
CF_3CN	95	W	b.p. -64	S	..
CF_2-CF_2 $N=N$	128	M	b.p. -125	P	..
CN-CN	52	W	b.p. -20.5	S	..
F F (ring with N)	135	W	m.p. -38, b.p. 73	P	..
CN C=O CN	80	S	m.p. -36, b.p. 66	S	R[d]
$CFO-C\equiv C-CFO$	118	S	b.p. 43	U	..

See footnotes at end of table.

TABLE 1. (Contd.)

Formula	Mol. wt.	Acceptor strength[a]	Volatility,[b] °C	Chemical stability[c]	Recom- mended
(F,F on central C; ring: C–CO–O–CO)	134	M	m.p. 20, b.p. 128	P	..
CF_2-CO $\mid \quad \mid$ CF_2-CO	156	S	b.p. 35	S on careful storage	R
$CF_3-C\equiv C-CF_3$	162	M	m.p. –118, b.p. –25	S	R
$CN-C\equiv C-CN$	76	S	m.p. 21, b.p. 77	S at low temp. and absence of oxygen	
$CN-C\equiv C-C\equiv C-CN$	100	S	m.p. 65, b.p. ca. 155	S absence of oxygen	..
$\begin{matrix} CN & & CN \\ & C{=}C & \\ CN & & CN \end{matrix}$	128	S	m.p. 197, s. 0.5 mm at 110	S	R

[a] W = weak, M = medium, S = strong.
[b] b.p. = boiling point, m.p. = melting point, s. = sublime.
[c] S = stable, P = probably stable (insufficient information), U = no information.
[d] See Appendix A.

As described in a previous article (Ref. 15), strong acceptors are for the most part highly unsaturated compounds included within the formula $C_w N_x O_y F_z$.

It is of interest that the same characteristics are also valuable in a chemical propellant. Electron affinity of a molecule is a measure of its oxidizing power. The specific impulse I of gases leaving a rocket nozzle is described approximately by the relation

$$ I \propto \sqrt{\frac{T}{M}} $$

where T is the temperature of the burning gases and M is their average molecular weight. Unsaturation, high-nitrogen proportions, N-O, N-F, O-O, and O-F bond systems confer high energy content and hence high T values on the fuel. For example, the strong acceptor dicyanoacetylene, on combustion, produces one of the hottest chemical flames known (Ref.

16 and 17). For small M values, it is necessary to use compounds of the lightest elements as fuels, and carbon, nitrogen, oxygen, and fluorine are within this category.

Unburnt liquid oxygen or liquid fluorine oxidizers are certainly expected to influence ionospheric electron densities. Even a very low percentage of electron accepting species (O_2, OH) in the exhaust of a U. S. Saturn rocket (100 tons above a height of 100 km) would be equivalent to n_e.

DELIVERY, DISPERSAL, AND DECAY

If the size of the payload is limited to 1 ton on the basis of the estimations in the first section of this report, and if equivalence of one molecule of scavenger per electron is assumed, compounds of molecular weight, M, up to 350 are acceptable for the volume 100 x 100 x 500 km^3. This includes all of the compounds in Table 1, although to limit the payload, the smallest possible values of M are of course preferred.

The scavenger will compete with other molecular species (Ref. 18) in the ionosphere for electrons. These include the strong acceptors N_2, O^+, and NO^+ from which the electrons have been photochemically detached and the weak acceptor O_2. Thus a scavenger concentration equal to the original electron concentration, although it may decrease n_e to below a critical value for radio-wave propagation or reflection, will only displace the equilibrium, depending on the strength of the acceptor. Photodetachment of electrons from the radical anions formed is certainly to be expected. For effective electron capture, concentrations up to an order of magnitude greater than n_e may have to be used. Another factor that will call for an increased load of scavenger will be diffusion of material away from the target area.

For certain interference with a long-range radar probe, the properties of the ionosphere must be altered over the total region where it is possible to reflect an enemy radio beam. Thus for 2,000-mile radar installations, screening of the ionosphere must be performed on a circle of 1,000 miles radius from an attacking site. As an example, if sea-based probes are neglected, an estimation for an exchange between England and Western Russia would require a 500-mile-wide belt of scavenging material, approximately 1,000 miles long, extending between Munich and Stockholm. This would involve a <u>hundred</u> times the quantities considered in the experiment.

Estimates for practical radar jamming are based on an acceptor, M 100, scavenging electrons, 2×10^{13} cm^{-2}, at a molecular concentration 2 n_e. The experimental volume will require 0.7 tons and the larger strategic

volume will require 70 tons. These quantities are within the delivery loads of present rockets, particularly as it will be advantageous to sub-divide the second, appropriate to the geometry of the screen.

One objection to this method of radar jamming is that the act of putting up the scavenger rocket will be detectable and will arouse suspicion. The rocket need not be directed towards enemy territory how-ever, or the material could be brought down on radio command, from an orbiting satellite. Both methods would favor the element of surprise. The attack weapon itself could be delivered from a satellite orbiting above the ionosphere, so obviating this method of radar detection. The geometry requirements of the orbit for a scavenger satellite would be far less precise than those for a weapon satellite.

High volatility, low critical temperature in particular, and small heats of vaporization are necessary for rapid dispersal of the scavenger. Low boiling liquids or readily sublimable solids in solution may be dis-charged as aerosols, although this process may add considerably to the weight. Alternatively, explosive dispersal by means of a charge at the center of the payload can be used for suitably stable materials. As reported above, strong acceptors are reactive electrophiles, Lewis acids, and oxidizing agents and for this reason they can be expected to react with water and many of the metals of containers.

Any practical means of dispersal will start with an initial high concentration of scavenger in a limited space. Diffusive expansion of this cloud will then occur into the projected volume. Release of 2-4 kg of alkali metal at the 125-km region has caused electron clouds to per-sist 30 minutes to 1 hour at night and even longer in the daylight, and this time is in agreement with a mathematical prediction (Ref. 4). Re-lease of 22 kg of sulfur hexafluoride at 220 km has caused (Ref. 19 and 20) an electron depleted region of 30-minute duration in daylight. (See Appendix B.) The day-night effect should be reversed for electron capture, whereas the higher concentrations proposed would cause more prolonged electron depletion at the positions of dispersal. Moreover the products of a high altitude nuclear explosion cause changes in radio-wave propagation up to periods of 2 weeks (Ref. 21).

"Holdup time" of radioactive fallout in the upper atmosphere and calculation (Ref. 4) indicate that the lighter scavenger materials will remain in the ionosphere for a year or more, although at an ineffective dilution. Repeated discharges over this period will be accumulative. Disintegration of the radical anions and acceptors by ultraviolet, X-radiation, and chemical reactions with constituents of the ionosphere will occur. At this high level though, it is likely that the products will still exhibit electron accepting properties. Thus, a possible mechanism for decomposition of a radical-anion from a halogen containing acceptor is the detachment of the halide ion from the molecule (e.g., F^-) to leave a reactive species that may undergo further decomposition or

may be able to accept a second electron. Final removal of material will result as some of the compounds, e.g., $(CN)_2CO$, undergo gradual decomposition in the presence of ultraviolet light and slowly diffuse back into the lower atmosphere, where harmless products at low concentrations will be formed from the interactions of the chemically reactive acceptors with water vapor.

CONCLUSIONS

Radar jamming on the scale proposed would favor an attacker, although it would render him vulnerable to immediate retaliation, assuming each adversary has the probe. As discussed above, this vulnerability would be short-lived, particularly if the quantity of scavenger is closely designed to requirements.

The estimated weights in the proposal are based on the 2×10^{13} electrons cm^{-2} figure, which is on the high and safe side, particularly for night-time conditions and temperate latitudes. Nevertheless, the quantities required for practical purposes are within the payloads of modern rockets.

Appendix A[2]

CARBONYL CYANIDE

Because of its acceptor properties, low molecular weight, 80, and low boiling point, 66°C, carbonyl cyanide is a valuable member of Table 1 for the purposes of investigation.

Hydrocarbons and other materials dissolve in it producing colors (Ref. 22). When carbon tetrachloride solutions of the cyanide and of the donors in Table 2 are mixed, single, broad, structureless, absorption bands, which are not present in either component separately, are produced immediately. These have the maxima shown, do not obey Beer's law, and appear reversibly on adding or diluting the components. Positions of the maxima correspond to changes in ionization potential of the donors and the bands are ascribed to charge-transfer transitions. Carbonyl cyanide is therefore a strong acceptor, stronger than trinitrobenzene or benzoquinone, but not as strong as chloranil or tetracyanoethylene. With respect to tetracyanoethylene, maxima for the complexes of the donors are displaced by 0.6 ev. Thus to a simple approximation, the electron affinity of carbonyl cyanide is smaller by this amount. Very approximately, its value is in the 3.0 ev region.

TABLE 2. Absorption of Complexes of Carbonyl Cyanide

Mesitylene	Durene	Hexamethylbenzene	Phenanthrene	Anthracene
375 mµ 3.30 ev	382 3.25	437 2.84	415[a] 2.99[a]	520 2.39

[a] Shoulder.

Prolonged action of ultraviolet radiation (Ref. 23) on the vapor decomposes the compound to carbon monoxide and the electron-accepting CN radical.

Sunlight is without marked effect on the stored liquid, although the slight darkening so produced has been minimized by the use of colored

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[2] This portion of the report is UNCLASSIFIED.

glass bottles. When all traces of moisture have been excluded, the material has been stored for sufficient periods to permit a prolonged investigation of its chemistry. Heating the cyanide to 100°C in the presence of active charcoal produces no decomposition.

Heat of combustion is high, 332 kcal/mole, and formation from the elements is endothermic, 50 kcal/mole (Ref. 24). Undoubtedly, care would be required to disperse the acceptor intact if explosive methods were used.

Carbonyl cyanide reacts violently with water, producing carbon dioxide and the exceedingly poisonous hydrogen cyanide. For the study of the complexes, it was prepared from acetone dicarboxylic acid by the method of Malachowski and Achmatowicz (Ref. 25 and 26). This method requires three stages, one of them dangerously exothermic, leading to a low over-all yield. Also careful and repeated distillation of the product is needed to remove traces of acetic acid, with which reaction occurs.

Further studies on this material are planned to examine other routes of preparation, to extend investigation of the complexes, to measure the polarographic reduction potential, and to make a prolonged examination of its thermal stability. A study of thermal electron capture (Ref. 27) will also be valuable.

Appendix 3

SCAVENGING RATES AND PROJECT FIREFLY

Scavenging Rates. Electron affinity of the acceptor is a measure of electron capture equilibrium and does not necessarily indicate the rate of scavenging. Strong radiative attachment requires a high probability of capture to an autoionizing level, together with a high probability of radiative dispersal of the energy to make the capture permanent. For the compounds in Table 1, very few measurements indeed have been made on electron capture cross sections, hence, little information is available on the probable rates of scavenging. A guess (Ref. 18) at the magnitude of the electron-scavenger recombination coefficient would be 10^{-8} cm^3 sec^{-1} for the combined radiative, nonradiative and dissociative attachment. At a concentration of 2 n_e of acceptor, where n_e is 10^6 electrons cm^{-3}, this would mean that approximately 2% of the electrons would be captured per second, or within 10 minutes, a turnover of 10 n_e could be accommodated. More precisely, electron scavenging according to the law of mass action relates electron concentration n at time t to the recombination coefficient k by the equation

$$n = \frac{n_e}{(2e^{n_e kt} - 1)}$$

and this indicates a half life of 40 seconds for the above figures.

An interesting possibility for the higher density D-layer of the ionosphere is to combine an acceptor of high capture cross section but of low electron affinity with an acceptor of low cross section and high electron affinity (the oxygen/nitrogen tetroxide system approximates to this situation). Here the speed of scavenging will depend on the rate that the second acceptor will remove anions of the first.

Another possibility is that a sudden release of even a neutral gas (N_2, CH_4) into the low pressure of the ionosphere should force electrons away from the point of ejection into the form of a high density surface of an expanding sphere.

Photoionization of atmospheric constituents is small over the period ($\sim$ 20 minutes) for which scavenging would be required and rates for the electronegative acceptors should be comparable. On the other hand, ample

sunlight of energy greater than the electron affinity of the acceptors (ca. 3 ev) is available to hamper daylight scavenging operations.

Project Firefly. Release of 22 kg of sulfur hexafluoride in the lower F-layer created an electron depleted region, effective diameter 20-30 km, detected by a ground-based radar and ionosonde, and this persisted up to 30 minutes (Ref. 19 and 20). A single-step electron attaching process was preferred for the project to one involving displacement of the atmospheric equilibrium towards electron removal (release of oxygen).

Sulfur hexafluoride was chosen because of the rapidity (Ref. 27) with which it attaches thermal electrons, 10^{-9} cm^3 sec^{-1}. However, this material has a high molecular weight, 146, and is a weak acceptor. An equivalent weight of carbonyl cyanide, 12 kg, may have a greater effect, should remain at the point of release in the ionosphere, and should have a more prolonged action as a result of photochemical decomposition to electron accepting CN radicals. On the other hand, it has to be dispelled as a low boiling liquid, instead of a gas heated, $65°C$, above its critical temperature.

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ABSTRACT CARD

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U. S. Naval Ordnance Test Station
Electron Acceptors as Electron Scavengers in the
Ionosphere (U), by P. R. Hammond. China Lake, Calif.,
NOTS, July 1965. 16 pp. (NAVWEPS Report 8764, NOTS
TP 3846), CONFIDENTIAL.
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